Impact of Nuclear Data Uncertainties on Advanced Fuel Cycles and their Irradiated Fuel - a Comparison between Libraries

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The uncertainties on the isotopic composition throughout the burnup due to the nuclear data uncertainties are analysed. The different sources of uncertainties: decay data, fission yield and cross sections; are propagated individually, and their effect assessed. Two applications are studied: EFIT (an ADS-like reactor) and ESFR (Sodium Fast Reactor). The impact of the uncertainties on cross sections provided by the EAF-2010, SCALE6.1 and COMMARA-2.0 libraries are compared. These Uncertainty Quantification (UQ) studies have been carried out with a Monte Carlo sampling approach implemented in the depletion/activation code ACAB. Such implementation has been improved to overcome depletion/activation problems with variations of the neutron spectrum.

I. INTRODUCTION

Propagation of nuclear data uncertainties in any nuclear system is necessary to determine properly the safety and economical margins. Even more, new reactor designs require an assessment of the accuracy of their main reactor parameters. In order to carry out this task, a complete set of covariance data should be provided, but also calculations tools that are capable of handling these data should be developed. With this framework, the European Framework Program 7 (FP7) is funding the ANDES project - Accurate Nuclear Data for nuclear Energy Sustainability.

Within this project, the ACAB code [1] is used to perform activation and burn-up calculations. New capabilities have been implemented to cope with nuclear data uncertainty propagation based on hybrid calculations [2]. Also, this methodology has been compared with others such as Total Monte Carlo [3], proving its capabilities. Then, advanced nuclear cycles can be analysed and the uncertainties on, for instance, isotopic composition, decay heat and radiotoxicity can be addressed.

Therefore, this paper is aimed to study the effect of different sources of uncertainty: decay data, fission yield and cross sections, in the fuel cycle of two advanced reactors: EFIT (European Facility for Industrial Transmutation) and ESFR (European Sodium Fast Reactor). The impact of the cross sections uncertainties of three different libraries: EAF-2010 [4], SCALE6.1 [5] and COMMARA-2.0 [6], are compared. The approach used for the Uncertainty Quantification (UQ) studies is based on a Monte Carlo sampling. The implementation in ACAB has been reviewed and improved, in order to cope with burnup problems where the spectrum changes during the irradiation time.

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II. REVISION OF UNCERTAINTY QUANTIFICATION APPROACH WITH ACAB

As shown in Ref. [2], the UQ approach using ACAB decouples transport and burn-up calculations, because ACAB can only deal with activation/burn-up problems. That means the uncertainties are only propagated to response functions that come from the depletion problem, mainly isotopic composition and its derivatives such as decay heat. However, coupling with transport uncertainties could be done by means of for instance sensitivity coefficients and the "Sandwich Formula" (Perturbation Theory).

With ACAB, the uncertainties in nuclear data are propagated by a Monte Carlo sampling. Thus means an amount of ACAB calculations is carried out with random cross-section, fission yield and decay data sampled with their uncertainty data. Each of these calculations is called a history. So, after running the whole amount of histories, a statistical analysis of the response functions such as isotopic composition, decay heat or radiotoxicity, is performed. Therefore, the uncertainties in nuclear data are propagated throughout the burn-up.

Burn-up calculations only require one-group cross sections, $\sigma_{1g,a}$, so the cross-section libraries are collapsed with the neutron spectrum, ϕ_i (Eq. 1). Usually multi-group structure libraries are used, σ_i, where i refers to the energy group. Their covariances, V, are also collapsed to one group (Eq. 2, derived from the Taylor expansions

for the moments of functions of random variables). With these data, random cross-section libraries can be sampled after assigning a Probability Density Function (PDF)

$$\sigma_{1g,a} = \sum_{i=1}^{n} \left(\frac{\phi_i}{\phi_T} \cdot \sigma_{i,a} \right) \quad (1)$$

$$var(\sigma_{1g,a}) = \omega^T V \omega \quad ; \quad \omega_i = \frac{\phi_i}{\phi_T}. \quad (2)$$

Because the neutron spectrum could change between burn-up steps, the cross-section mean values, $\sigma_{1g,a}$, and their variance-covariance would be modified. So, a different reference library for sampling would be used for each step. However, the original random variables were the multi-group cross sections, $\sigma_{i,a}$, and they do not change between burn-up steps. If the multi-group cross-sections were sampled instead, the random cross section would be determined from the very beginning because in each step the random one-group cross-section would be calculated collapsing the random multi-group cross section with the neutron spectrum of the burn-up step. Hence, if one-group random cross-sections are used, that implies a relationship between the one-group random cross section values of each burn-up step for a given history.

This relationship is implemented by using the same random number to sampled the one-group cross-section in every burn-up step. In this way, the random number used to define a one-group random cross-section is equivalent to the set of random multi-group cross-section.

Other methodologies, e.g. Total Monte Carlo [7] or XSUSA [8], that used nuclear parameters or multi-group libraries, are not affected by this issue because the original random variables are not converted into others.

III. ACTIVATION/BURN-UP CALCULATIONS IN ADVANCED FUEL CYCLES

The fuel cycle of two different advanced reactors are studied: EFIT and ESFR. For both cases, EVOLCODE-2.0 [9] is used to calculate the neutron flux and spectrum throughout the burnup with the ENDF/B-VII.0 library [10]. After, the UQ studies on the isotopic composition of a representative fuel cell during the burnup due to the nuclear data uncertainties are performed. The uncertainties taken into account come from cross-sections, fission yields and decay data.

For decay and fission yield data, uncertainties are taken from JEFF-3.1.1 [11] for both cases. The uncertainties are propagated individually, in order to show their solely effect. Finally, to ensure convergence, 1000 histories are launched for each kind of uncertainty propagated.

A. EFIT

EFIT is a ADS-like reactor, aimed to burning Minor Actinides (MAs), with a neutron flux intensity of 3.12×10^{15} n/cm^{2}s and a fast spectrum ($\bar{E}$=0.37514 MeV). As design point, a 21.7% of MAs are charged with fresh fuel, and runs at 400 MW for 778 days in order to achieve a burnup of 150 GWd/tHM. This configuration was previously studied [12]. However, here it is updated with the propagation of the uncertainties in EAF-2010 [4].

The isotopes whose uncertainty changes because of using EAF-2010 [4] instead of EAF-2007 [13] are presented in Table I. There, the uncertainties on the number of atoms at the discharge are shown for each source of uncertainty. In general, the uncertainties in almost every isotope decrease. Only the ^{240}Pu and ^{242}Cm uncertainties increase.

TABLE I. Uncertainties on the number of atoms for the most relevant changes obtained from using EAF-2010 instead of EAF-2007 at the discharge burn-up of 150 GWd/tHM for EFIT.

Nuclide	Uncertainty (as rel. std. dev. %)				
	Decay	FYs	Cross-section		
	JEFF-3.1.1		EAF-2007	SCALE	EAF-2010
^{235}U	0	-	13.2	3.0	3.6
^{237}Np	0	-	6.1	1.4	3.0
^{240}Pu	0	-	1.9	0.3	3.2
^{241}Pu	0	-	8.3	0.9	2.9
^{241}Am	0	-	7.0	2.0	3.5
^{243}Am	0	-	6.1	1.4	2.7
^{242}Cm	0.1	-	10.4	3.4	14.0
^{243}Cm	0.2	-	23.4	11.7	3.7
^{245}Cm	0	-	13.2	9.7	3.4
^{247}Cm	0	-	15.7	11.0	6.3
^{249}Bk	1	-	20.2	17.3	7.4
^{252}Cf	0.3	-	56.4	35.6	15.4
^{94}Nb	0.03	5.9	17.6	4.6	4.3
^{93}Mo	0.01	2.7	82.6	1.2	35.7
^{126}Sb	5.21	9.2	9.0	3.3	2.9
^{126M}Sb	1.05	7.5	16.4	1.9	6.6
^{150}Sm	0.01	3.0	11.0	7.7	5.2
^{151}Sm	0.05	4.2	10.9	6.7	3.1

B. ESFR

For this advanced reactor design [14], the uncertainties in the isotopic composition of a characteristic fuel cell at the end of the irradiation stage is calculated. The burn-up (99 GWd/tHM) is divided into five steps to take into account the variation of the flux intensity and the spectrum (see Table II). The neutron spectrum seen by the studied cell is presented in Fig. 1, where the spectra at the Beginning of Cycle (BOC) and at the End of Cycle (EOC) are presented.

For the oncoming calculations, cross section data are retrieved from EAF-2010 [4]. However, due to the aim of comparing the effect of cross section uncertainties from different libraries, the uncertainties stored in EAF-2010 are substituted with those in SCALE6.1 [5] and

TABLE II. ESFR Neutron flux intensity during the burnup.

Time(days)	Flux intensity (n/cm^{2}s)
410	3.1637×10^{15}
820	2.9798×10^{15}
1230	3.1637×10^{15}
1640	2.5980×10^{15}
2050	2.4464×10^{15}

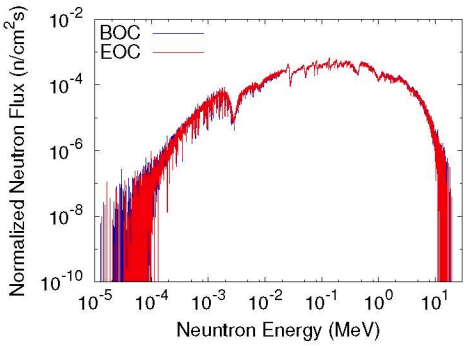

FIG. 1. ESFR neutron spectra at BOC and EOC.

COMMARA-2.0 [6], when the effects of these last two are assessed. As suggested in Ref. [5], the relative standard deviation values are used regarding the uncertainties.

Because the burn-up is split into five burn-up steps, thanks to the revision in Section II this problem can be overcome (Case A). Although, two more approaches are carried out:

- Not correlating the random cross-section between burn-up steps for a given history (Case B).
- Using the random cross-sections sampled in the first step for every step (Case C).

Case A and Case C should provide very similar values because the neutron spectrum almost does not change from BOC to EOC. Meanwhile, Case B approximation is not consistent because the isotopic composition should be sampled after every burn-up step if the random cross-section are not correlated between steps.

The results are compiled in Table III, where the initial composition and its variation between the beginning and end of burn-up, and the uncertainties on the number of atoms obtained from the different sources/cases are shown. As expected, a good agreement between Case A and Case C is observed, while Case B provides a completely random behaviour, as shown in Fig. 2, that provokes an underestimation of the uncertainties except for ^{242}Cm, ^{126}Sb and ^{126m}Sb.

Decay data uncertainties are only of relevance for two fission products:^{126}Sb and ^{151}Eu. For ^{151}Eu, its uncertainty comes from the 6.67% uncertainty on the ^{151}Sm decay constant, because the ^{151}Sm decay is the main production source of ^{151}Eu. However, the removal of ^{151}Sm is ruled by cross sections, not by decay. So that, the effect of its decay uncertainty is almost negligible, and then, a very small uncertainty value is achieved on the ^{151}Sm concentration.

Large differences are found between using uncertainties from different cross-section libraries. EAF-2010 produces higher uncertainties than SCALE and COMMARA-2.0 except for isotopes heavier than ^{242}Cm, while these last two have a similar performance.

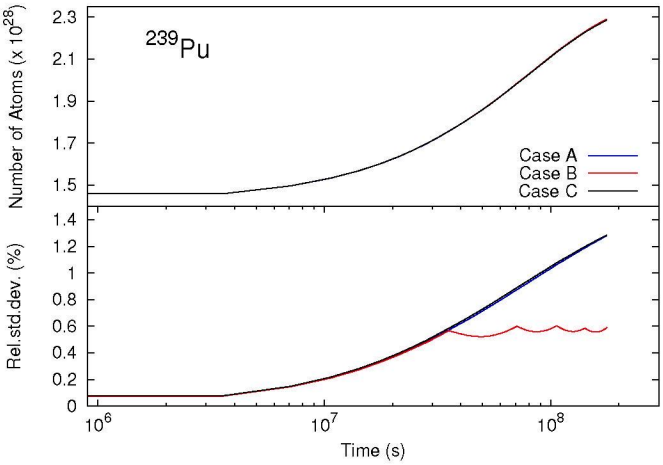

FIG. 2. Number of atoms of ^{239}Pu throughout the burn-up and its uncertainty for each of the cases studied using EAF-2010+SCALE uncertainties.

IV. CONCLUSIONS

An improvement of the Uncertainty Quantification (UQ) approach implemented in ACAB is performed in order to deal with burn-up/activation problems where there are variations of neutron spectrum between burn-up steps.

The UQ study for EFIT is updated using the EAF-2010 library, obtaining smaller uncertainties than previously. For ESFR, the new implementation of the ACAB methodology is used and compared with other two approaches to check that the problem is properly overcome. The effect of the cross section uncertainties in EAF-2010, SCALE and COMMARA-2.0 are compared, showing that EAF-2010 provides a higher uncertainty for isotopes heavier than ^{242}Cm, while SCALE and COMMARA-2.0 provide very similar results.

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TABLE III. Uncertainties on the atomic composition of heavy isotopes and fission products for a ESFR characteristic fuel cell after 99 GWd/tHM burn-up.

Nuclide	N_i (atoms)	N_f - N_i (atoms)	Uncertainty (as rel.std.dev. %)										
			Decay	FYs	EAF-2010			SCALE			COMMARA-2.0		
			JEFF-3.1.1		Case A	Case B	Case C	Case A	Case B	Case C	Case A	Case B	Case C
^{235}U	5.24×10^{26}	-3.52×10^{26}	0.01	-	6.56	2.86	6.41	6.19	2.85	6.29	4.02	1.69	3.82
^{238}U	2.04×10^{29}	-3.28×10^{28}	0.00	-	0.64	0.29	0.63	0.23	0.11	0.24	0.22	0.10	0.21
^{237}Np	1.65×10^{27}	-8.29×10^{26}	0.00	-	6.32	2.86	6.19	3.19	1.39	3.19	3.25	1.47	3.21
^{238}Pu	1.08×10^{27}	1.35×10^{27}	0.04	-	7.62	3.60	7.57	4.32	2.10	4.41	3.32	1.56	3.53
^{239}Pu	1.43×10^{28}	7.65×10^{27}	0.00	-	5.29	2.48	5.17	1.28	0.59	1.29	1.30	0.61	1.21
^{240}Pu	8.89×10^{27}	6.52×10^{26}	0.00	-	3.22	1.47	3.21	2.15	1.01	2.12	2.39	1.06	2.30
^{241}Pu	2.46×10^{27}	-7.78×10^{26}	0.02	-	5.75	2.78	6.11	1.70	0.83	1.67	3.69	1.83	3.58
^{242}Pu	3.09×10^{27}	-3.75×10^{26}	0.05	-	2.84	1.32	2.79	1.29	0.53	1.32	3.87	1.73	3.92
^{241}Am	5.84×10^{27}	-3.38×10^{27}	0.02	-	12.39	5.92	12.47	3.86	1.72	3.83	1.79	0.86	1.85
^{242m}Am	2.30×10^{25}	7.60×10^{25}	0.01	-	18.69	8.99	18.50	18.95	8.43	17.91	18.37	8.77	19.60
^{243}Am	1.50×10^{27}	-4.04×10^{26}	0.02	-	3.19	1.45	3.37	4.11	1.88	4.02	6.98	3.10	7.02
^{242}Cm	1.92×10^{24}	1.67×10^{26}	0.12	-	6.42	13.43	6.48	1.91	3.79	1.84	3.39	3.07	3.48
^{243}Cm	6.69×10^{24}	2.05×10^{25}	0.22	-	16.00	7.89	16.08	19.41	8.95	18.45	54.79	25.07	52.79
^{244}Cm	4.89×10^{26}	3.98×10^{26}	0.04	-	3.09	1.53	3.13	5.76	2.60	5.81	9.12	4.43	9.15
^{245}Cm	1.19×10^{26}	3.80×10^{25}	0.02	-	6.99	3.49	7.12	16.21	7.91	16.31	35.08	17.96	34.27
^{246}Cm	8.49×10^{24}	2.49×10^{25}	0.01	-	5.99	2.61	5.89	13.09	6.08	13.46	21.84	10.12	21.42
^{247}Cm	-	3.11×10^{24}	0.00	-	8.55	4.52	8.30	24.10	13.24	25.12	26.11	14.94	27.63
^{248}Cm	-	3.46×10^{23}	0.00	-	10.07	5.02	10.20	33.59	17.26	34.41	25.05	13.04	26.57
^{249}Bk	-	5.27×10^{21}	0.91	-	10.72	6.28	10.82	42.35	23.75	41.78	25.12	14.08	26.38
^{252}Cf	-	1.00×10^{18}	0.21	-	15.65	9.66	15.68	58.97	33.56	55.04	27.83	18.98	29.09
^{93m}Nb	-	1.27×10^{21}	0.09	13.12	5.66	2.79	5.66	1.09	0.63	1.13	1.02	0.63	1.08
^{94}Nb	-	7.34×10^{21}	0.00	9.38	12.58	5.53	12.84	7.72	3.49	7.57	7.87	3.45	7.71
^{93}Mo	-	8.91×10^{18}	0.01	14.63	30.22	18.17	31.21	27.05	16.08	27.86	29.81	17.82	31.20
^{103}Rh	-	1.64×10^{27}	0.00	2.53	5.08	2.22	5.10	1.08	0.57	1.11	1.02	0.51	0.99
^{107}Pd	-	7.80×10^{26}	0.00	2.43	4.58	2.08	4.60	2.20	1.23	2.29	2.32	1.18	2.25
^{109}Ag	-	4.08×10^{26}	0.00	2.65	5.51	2.55	5.72	2.17	1.07	2.14	2.20	1.08	2.18
^{126}Sn	-	6.24×10^{25}	0.00	4.01	4.80	2.15	4.87	0.81	0.38	0.81	0.74	0.36	0.70
^{126}Sb	-	3.51×10^{22}	5.89	19.81	4.80	6.73	4.82	6.98	7.18	6.59	1.39	1.12	1.29
^{126m}Sb	-	6.13×10^{19}	0.99	13.43	6.17	7.02	6.05	3.92	3.92	3.91	4.22	4.09	3.98
^{129}I	-	3.36×10^{26}	0.03	2.36	4.79	2.16	4.86	1.34	0.69	1.46	0.82	0.41	0.79
^{149}Sm	-	2.68×10^{26}	0.00	2.85	5.47	2.56	5.67	5.36	2.79	5.18	5.59	2.74	5.19
^{150}Sm	-	1.31×10^{26}	0.00	3.03	7.06	3.55	6.90	10.53	5.62	10.39	10.68	5.31	9.91
^{151}Sm	-	1.46×10^{26}	0.06	2.74	5.08	2.43	5.09	9.49	4.97	9.20	7.93	4.31	7.37
^{152}Sm	-	2.46×10^{26}	0.01	1.97	4.97	2.22	5.05	5.00	2.66	4.91	4.51	2.49	4.27
^{151}Eu	-	2.19×10^{24}	3.85	2.76	6.70	3.62	6.65	10.69	5.87	10.27	7.76	3.94	7.31
^{153}Eu	-	8.48×10^{25}	0.01	2.54	12.08	6.17	12.05	6.75	3.61	6.62	6.79	3.60	6.65
^{155}Gd	-	1.30×10^{25}	0.13	3.04	6.53	3.10	6.40	4.27	2.23	4.23	4.31	2.23	4.33

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